Tunneling with dissipation and decoherence for a large spin.

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We present rigorous solution of problems of tunneling with dissipation and decoherence for a spin of an atom or a molecule in an isotropic solid matrix. Our approach is based upon switching to a rotating coordinate system coupled to the local crystal field. We show that the spin of a molecule can be used in a qubit only if the molecule is strongly coupled with its atomic environment. This condition is a consequence of the conservation of the total angular momentum (spin + matrix), that has been largely ignored in previous studies of spin tunneling.

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The problem of tunneling of a large spin [1,2,3,4,5,6,7,8,9] has received considerable attention lately in connection with spin-10 magnetic molecules, Mn-12 and Fe-8, [10,11,12,13,14,15,16,17]. High-spin molecules have been proposed as qubits for quantum computers [18]. It is, therefore, important to understand the effect of the atomic environment on spin tunneling and decoherence. In this Letter we present rigorous solution of both problems for a spin of a molecule in an isotropic solid.

The Caldeira-Leggett approach [19] to the problem of spin tunneling with dissipation due to phonons was outlined in the second of Refs. [4]. It has been applied in Ref. [20], though a general expression for the effective action has not been obtained. The correct formulation of the problem should account for the conservation of the total angular momentum: spin $\mathbf{S}$ plus the angular momentum $\mathbf{L}$ of the atomic lattice [21]. In the absence of the external field, tunneling of $\mathbf{S}$ between $\Uparrow$ and $\Downarrow$ should be accompanied by a simultaneous co-flipping of $\mathbf{L}$ and is possible only if $\mathbf{J} = \mathbf{S} + \mathbf{L} = 0$. This can be satisfied by an integer but not by a half-integer S, which is another way to look at the Kramers theorem. The necessity to have $L = S$ in the tunneling state results in the mechanical rotational energy $E_r = (\hbar S)^2/2\mathcal{I}$ where $\mathcal{I} \sim \rho l^5$ is the moment of inertia of the atomic lattice; ρ and l being the mass density and the linear dimension of the solid matrix containing $\mathbf{S}$. This energy should be compared with the tunneling splitting Δ. The condition $\Delta > E_r$ is needed for the tunneling state with $L = S$, $J = 0$ to be the ground state of the system, while in the opposite case of $\Delta < E_r$ the ground state should be $L = 0$, $J = S$, with $\mathbf{S}$ frozen along the anisotropy axis. This translates into a minimal size l (typically of order 1-10 nanometers) of a free particle whose spin can tunnel between equilibrium orientations.

The problem of the decoherence is even more subtle. On one hand, the matrix elements of the conventional spin-lattice interaction due to the electrostatic crystal field vanish between $(|\Uparrow> + |\Downarrow>)$ and $(|\Uparrow> - |\Downarrow>)$ tunneling spin states [22]. On the other hand, the real-time coherent oscillations of $\mathbf{S}$ in a solid matrix must be accompanied, through the conservation of the angular momentum, by the oscillating shear deformation of the solid. This should be the source of decoherence. The relevant size of the solid involved in that process is $l_c = c_t/\omega_c$, where c_t is the velocity of the transverse sound and $\omega_c = \Delta/\hbar$ is the oscillation frequency. We shall see that $\Gamma = E_r/\hbar \sim \hbar S^2/\rho l_c^5$ emerges as a frequency scale that determines the decoherence rate.

The imaginary-time action of the system consists of the action of the spin in a crystal field of the atomic lattice and the action of the lattice. The spin action can be written as

$$I_s[\mathbf{n}] = I_{WZ}[\mathbf{n}] + \int_0^{\hbar\beta} d\tau E_s(\mathbf{n}) , \qquad (1)$$

where I_{WZ} is the Wess-Zumino action [9], E_s is the energy of the magnetic anisotropy due to the crystal field, $\mathbf{n}(\tau) = \mathbf{S}(\tau)/S$, and $\beta = 1/T$. The anisotropy is determined by the local atomic environment of the atom or molecule with spin $\mathbf{S}$. The global symmetry of the lattice is less important since the wavelengths involved in the problem (see below) are always large compared to the atomic scale. For that reason, and in order to simplify calculations, it is convenient to choose an isotropic (e.g., amorphous) solid that is characterized by two elastic moduli only, μ and λ, which determine velocities of the transverse and longitudinal sound, $c_t = (\mu/\rho)^{1/2}$, $c_l = [(\lambda + 2\mu)/\rho]^{1/2}$. Within linear elastic theory the corresponding phonon action is

$$I_l[\mathbf{u}] = \int_0^{\hbar\beta} d\tau \int d^3\mathbf{r} \left\{ \frac{1}{2}\rho\dot{\mathbf{u}}^2 + \mu(u_{ij})^2 + \frac{\lambda}{2}u_{kk}^2 \right\} , \quad (2)$$

where $\mathbf{u}(\tau)$ is the phonon displacement field and $u_{ij} = \frac{1}{2}(\partial_i u_j + \partial_j u_i)$ is the strain tensor.

We shall assume throughout this Letter that the interaction of the spin with its atomic environment is contained *entirely* in the structure of the crystal field responsible for the magnetic anisotropy. For example, in Mn-12 the spin Hamiltonian is dominated by the uniaxial anisotropy, $\mathcal{H}_s = -D(\mathbf{e}\cdot\mathbf{S})^2$ where $\mathbf{e}$ is the unit vector along the anisotropy axes. The conventional way to introduce the spin-phonon interaction [11,14] is to consider a small perturbation of $\mathbf{e}$ by phonons, $\delta\mathbf{e} = [\delta\phi\times\mathbf{e}]$, where $\delta\phi = \frac{1}{2}\nabla\times\mathbf{u}$ is the local rotation. This results in the spin-phonon Hamiltonian

$$\mathcal{H}_{sp} = D\{S_z, S_x\}\omega_{zx} + D\{S_z, S_y\}\omega_{zy} , \qquad (3)$$

where $\{S_i, S_j\}$ is the anti-commutator and $\omega_{ij} = \frac{1}{2}(\partial_i u_j - \partial_j u_i)$. The effective spin action,

$$I_{eff}[\mathbf{n}] = I_s[\mathbf{n}] + I_{env}[\mathbf{n}] , \qquad (4)$$

should be obtained by computing the environmental action I_{env},

$$\exp\left(-\frac{I_{env}[\mathbf{n}]}{\hbar}\right) = \frac{1}{Z_l}\oint \mathcal{D}\mathbf{u}\exp\left(-\frac{I_l[\mathbf{u}]}{\hbar} - \frac{I_{int}[\mathbf{n},\mathbf{u}]}{\hbar}\right) , \qquad (5)$$

where

$$Z_l = \oint \mathcal{D}\mathbf{u}(\mathbf{r},\tau)\exp\left(-\frac{I_l[\mathbf{u}]}{\hbar}\right) . \qquad (6)$$

The conventional approach [4,20] is to choose $I_{int} = \int_0^{\hbar\beta} d\tau \mathcal{H}_{sp}(\mathbf{n})$ with $\mathcal{H}_{sp}$ of Eq.(3) or similar. Then the integration in Eq.(5), though cumbersome, is Gaussian and, in principle, can be performed exactly. As has been discussed above, the problem with such an approach is that the tunneling of $\mathbf{S}$ from the $\Uparrow$ state at $\tau = -\infty$ to the $\Downarrow$ state at $\tau = +\infty$ must formally involve the co-flipping of $\mathbf{L}$, that is, the change of the mechanical rotation of the solid from clockwise to counterclockwise. In a fixed coordinate system the corresponding instanton involves large dispacements $\mathbf{u}$ which are difficult to work with.

The difficulty mentioned above can be avoided by switching to a coordinate system that is centered at the spin and is firmly coupled to the local anisotropy axes. It rotates in the presence of the time-dependent shear deformation. In such a coordinate system the magnetic anisotropy $E_s[\mathbf{n}]$ remains unaffected by phonons and the tunneling of the spin is accompanied by small lattice displacements only. The new term appears in the energy, though,

$$E'_s = -\hbar\mathbf{S}\cdot\mathbf{\Omega} , \qquad (7)$$

where $\mathbf{\Omega} = \delta\dot\phi = \frac{1}{2}\nabla\times\dot{\mathbf{u}}$. This can be considered as the consequence of the fact that rotation is equivalent to the magnetic field. In the rotating coordinate system I_{int} in Eq.(5) becomes

$$I_{int} = -i\hbar S\int_0^{\hbar\beta} d\tau\dot{\mathbf{n}}\left[\frac{1}{2}\nabla\times\mathbf{u}\right]_{\mathbf{r}=0} , \qquad (8)$$

where we have integrated by parts to move the time derivative to $\mathbf{n}$. Consequently, Eq.(5) is again a simple Gaussian integral on phonon variables.

Equations (7) and (8) can be mistakenly taken for a parameter-free spin-phonon interaction [11]. It is important to understand, therefore, that they are solely the consequence of switching to a rotating coordinate system, where the interaction between $\mathbf{S}$ and the lattice is contained, to all orders on $\mathbf{u}$, in the anisotropy energy $E_s[\mathbf{n}]$. To illustrate the equivalence of this approach to the conventional method of studies of the spin-phonon interaction, let us compute, e.g., the width, Γ_1, of the $T = 0$ spin-precession resonance for $\mathcal{H}_s = -DS_z^2$, that is the rate of the one-phonon decay $|m = S-1> \rightarrow |m = S>$, provided by Eq.(3) and by Eq.(7). After standard quantization of $\mathbf{u}$ (see below for details) Eq.(3) gives [23,11,14] $\Gamma_1 = S(2S-1)^2 D^2\omega_1^3/12\pi\hbar\rho c_t^5$, while Eq.(7) gives $\Gamma'_1 = S\hbar\omega_1^5/12\pi\rho c_t^5$, where $\hbar\omega_1$ is the distance between the $m = S$ and $m = S-1$ levels. Observing that this distance equals $(2S-1)D$, one immediately obtains $\Gamma_1 = \Gamma'_1$. The equivalence of the two methods in general can be traced to the equation of motion for the spin operator, $\hbar(d\mathbf{S}/d\tau) = [\mathcal{H}, \mathbf{S}]$.

We shall now proceed to the computation of the Caldeira-Leggett action. In terms of phonon modes,

$$\mathbf{u}(\mathbf{r},\tau) = \frac{1}{\sqrt{N}}\sum_{\mathbf{k}\lambda} e^{i\mathbf{k}\cdot\mathbf{r}}\mathbf{e}_{\mathbf{k}\lambda}u_{\mathbf{k},\lambda}(\mathbf{r},\tau) , \qquad (9)$$

I_l and I_{int} become

$$I_l[\mathbf{u}] = \int_0^{\hbar\beta} d\tau\sum_{\mathbf{k}\lambda}\frac{1}{2}M(\dot u_{-\mathbf{k}\lambda}\dot u_{\mathbf{k}\lambda} + \omega_{\mathbf{k}\lambda}^2 u_{-\mathbf{k},\lambda}u_{\mathbf{k}\lambda})$$

$$I_{int}[\mathbf{u},\mathbf{n}] = -i\int_0^{\hbar\beta} d\tau\sum_{\mathbf{k}\lambda}\dot{\mathbf{n}}\cdot\mathbf{C}_{\mathbf{k}\lambda}u_{\mathbf{k}\lambda} , \qquad (10)$$

where

$$\mathbf{C}_{\mathbf{k}\lambda} = \frac{1}{2\sqrt{N}}\hbar S(i\mathbf{k})\times\mathbf{e}_{\mathbf{k}\lambda} , \qquad (11)$$

$\mathbf{e}_{\mathbf{k},\lambda}$ are phonon polarization vectors, $\mathbf{e}_{-\mathbf{k},\lambda}\cdot\mathbf{e}_{\mathbf{k}\lambda'} = \delta_{\lambda\lambda'}$, M is the unit cell mass, and N is the number of unit cells in the lattice. The result of the integration over phonon variables in Eq.(5) can be presented as a sum of the local and nonlocal terms,

$$I_{env}[\mathbf{n}] = I_{env}^l[\mathbf{n}] + I_{env}^{nl}[\mathbf{n}] , \qquad (12)$$

where the local part is given by

$$I_{env}^l[\mathbf{n}] = \frac{\hbar^2 S^2}{12\mu V_c}\int_0^{\hbar\beta} d\tau\dot{\mathbf{n}}^2 , \qquad (13)$$

and the nonlocal part is

$$I_{env}^{nl} = -\frac{9\hbar^2 S^2}{32\pi\mu c_t^3}\int_0^{\hbar\beta} d\tau$$
$$\int_{-\infty}^{\infty} d\tau' f(\omega_D^t|\tau - \tau'|)\frac{[\dot{\mathbf{n}}(\tau) - \dot{\mathbf{n}}(\tau')]^2}{|\tau - \tau'|^4} . \qquad (14)$$

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Here $V_c = M/\rho$ is the unit cell volume, $\omega_D^t = c_t/V_c^{1/3}$ is the Debye frequency for the transverse phonon branch, and

$$f(x) = \frac{1}{6} \int_0^x dx' x'^3 e^{-x'} .$$ (15)

The typical scale of the time derivative in Eqs. (13) and (14) is set by the instanton frequency, ω_i, which also determines the temperature, $T_c \sim \hbar \omega_i$, of the crossover from superparamagnetism to quantum tunneling of **S**. The scale of T_c is set by E_s/S [9]. It is easy to see that $|I_{env}^{nl}/I_{env}^l| \sim (T_c/T_D)^2$. This ratio is small as long as the energy of the magnetic anisotropy is small compared to the elastic energy, which is normally the case. The relative effect of the atomic environment on spin tunneling is given by the ratio I_{env}/I_s, which is of the order of $E_s/\mu V_c$. For a molecule of spin **S**, *rigidly* imbedded in a solid matrix, this ratio is typically $10^{-6} - 10^{-3}$. On the contrary, *a loose* coupling of the magnetic molecule with its atomic environment is equivalent to $\mu \to 0$, which must result in a drastic increase of I_{eff} and the corresponding exponential decrease of the tunneling rate.

The advantage of our method becomes apparent when one wants to compute the rate of the decoherence for quantum oscillations of **S** between $\Uparrow$ and $\Downarrow$. Here we assume that the tunneling splitting Δ has been already renormalized by the effects studied above. If Δ is small, the problem can be trancated to the spin 1/2 problem with only two states: the ground state $|0> = \frac{1}{\sqrt{2}}(|\Uparrow> + |\Downarrow>)$ and the excited state, $|1> = \frac{1}{\sqrt{2}}(|\Uparrow> - |\Downarrow>)$. At $T = 0$ the decoherence occurs due to the transition $|1> \to |0>$ via spontaneous emission of a phonon of energy $\Delta = \hbar \omega_c$. Such transitions seem to be absent for the conventional magneto-elastic coupling that one obtains by considering the *adiabatic* rotation of the electrostatic crystal field (see, e.g., Eq.(3)). To compute the decoherence rate, one has to take into account the *dynamical* nature of the phonon [24]. This can be done by switching to a rotating coordinate system coupled to the local crystal field.

The general formula for the transition rate is

$$\Gamma = \frac{2\pi}{\hbar} \sum_{i \neq j} <i|\hat{V}|j><j|\hat{V}|i> \delta(E_i - E_j) .$$ (16)

Substituting here $\hat{V} = -\hbar \hat{\mathbf{S}} \cdot \hat{\boldsymbol{\Omega}}$, we get

$$\Gamma = 2\hbar <0|\hat{\mathbf{S}}|1> [J(\Delta)] <1|\hat{\mathbf{S}}|0> ,$$ (17)

where $J(\Delta)$ is the spectral function of the environmental coupling,

$$J(\Delta) = \pi \sum_{\mathbf{k}\lambda} <\mathbf{k}\lambda|\hat{\boldsymbol{\Omega}}|0><0|\hat{\boldsymbol{\Omega}}|\mathbf{k}\lambda> \delta(\Delta - \hbar \omega_{\mathbf{k}\lambda}) .$$ (18)

Computing the spin matrix elements, one gets

$$\Gamma = 2\hbar S^2 J(\Delta) .$$ (19)

To compute the spectral function, let us consider

$$\mathcal{J}(\Delta) = \pi \sum_{\mathbf{k}\lambda} \boldsymbol{\Omega}_{\mathbf{k}\lambda}^* \otimes \boldsymbol{\Omega}_{\mathbf{k}\lambda} \delta(\Delta - \hbar \omega_{\mathbf{k}\lambda}) ,$$ (20)

where

$$\boldsymbol{\Omega}_{\mathbf{k}\lambda} = <0|\hat{\boldsymbol{\Omega}}|\mathbf{k}\lambda> .$$ (21)

Here

$$\hat{\boldsymbol{\Omega}} = \frac{1}{2}[\nabla \times \dot{\mathbf{u}}]_{r=0} = \frac{1}{2\rho}[\nabla \times \hat{\boldsymbol{\Pi}}(\mathbf{r},t)]_{r=o}$$ (22)

and $\hat{\boldsymbol{\Pi}}(\mathbf{r},t)$ is the momentum of phonons in terms of the operators of creation and annihilation,

$$\hat{\boldsymbol{\Pi}}(\mathbf{r},t) = -\frac{i}{\sqrt{V}} \sum_{\mathbf{k}\lambda} \sqrt{\frac{\hbar \omega_{\mathbf{k}\lambda}}{2\rho}} [\hat{a}_{\mathbf{k}\lambda} \mathbf{e}_{\mathbf{k}\lambda} e^{i\mathbf{k}\cdot\mathbf{r}} - h.c.] .$$ (23)

This gives

$$\boldsymbol{\Omega}_{\mathbf{k}\lambda} = -i\sqrt{\frac{\hbar \omega_{\mathbf{k}\lambda}}{2M}} \mathbf{c}_{\mathbf{k}\lambda} , \quad \mathbf{c}_{\mathbf{k}\lambda} = \frac{1}{2\sqrt{N}} i\mathbf{k} \times \mathbf{e}_{\mathbf{k}\lambda} .$$ (24)

Consequently, Eq.(20) becomes

$$\mathcal{J}(\Delta) = \frac{\pi}{2} \sum_{\mathbf{k}\lambda} \frac{\Delta}{M} \mathbf{c}_{\mathbf{k}\lambda}^* \otimes \mathbf{c}_{\mathbf{k}\lambda} \delta(\Delta - \hbar \omega_{\mathbf{k}\lambda}) .$$ (25)

The spectral function is $J = \frac{1}{3} Tr[\mathcal{J}(\Delta)]$, which gives

$$J(\Delta) = \frac{\pi}{6} \sum_{\mathbf{k}\lambda} \left(\frac{\Delta}{M}\right) \left(\frac{k^2}{4N}\right) \delta_{\lambda t} \delta(\Delta - \hbar \omega_{\mathbf{k}\lambda}) .$$ (26)

The summation over phonon modes yields

$$J(\Delta) = \frac{\Delta^5}{24\pi \hbar^5 \rho c_t^5} .$$ (27)

Substituting Eq.(27) into Eq.(19) we finally obtain

$$\Gamma = \frac{\hbar S^2}{12\pi \rho l_c^5}$$ (28)

for the decoherence rate. Here we have introduced $l_c = c_t/\omega_c$. The generalization to finite temperatures is trivial. It results in the multiplication of the zero-temperature Γ of Eq.(28) by the factor $\coth(\Delta/2T)$. Note that the form of Eq.(28) is independent of the form of the spin Hamiltonian as long as the crystal field is the only source of the spin-lattice coupling. The dependence of Γ on the parameters of the crystal field is absorbed into $\Delta = \hbar \omega_c$.

The physics behind Eq.(28) is this [25]. The real-time quantum oscillations of **S** produce oscillating torque in the surrounding matter. That torque results in the torsion oscillations of the solid adjacent to the magnetic molecule, so that $\mathbf{J} = \mathbf{S} + \mathbf{L} = 0$. The corresponding

time-dependent elastic deformation is a coherent quantum superposition of diverging and converging sound waves. That deformation is confined within distances of order $l_c = c_t/\omega_c$ from **S**. By order of magnitude, l_c determines how far the sound goes away from the spin during the period of coherent quantum oscillations. At times of order Γ^{-1} the coherent superposition of sound waves is destroyed by a diverging wave of energy Δ that escapes to the surface of the solid. The energy $\hbar\Gamma$ determines the width of the excited state $|1>$. It is easy to see that $\hbar\Gamma \sim L^2/2\mathcal{I}_c$, where $L = \hbar S$ is the angular momentum associated with the spin and $\mathcal{I}_c \sim \rho l_c^5$ is the moment of inertia of the volume of spatial dimensions l_c.

If the magnetic molecule is to serve as a qubit, the frequency ω_c should be sufficiently high, in the ballpark of $10^{10} - 10^{11} s^{-1}$. Our study shows that it is possible only if the molecule is rigidly coupled to a solid matrix. This is contrary to the widely accepted view that the decoupling from the environment should be beneficial for the work of the qubit. In fact, *any effort to make a loose connection of the spin with its atomic environment, by e.g. having a magnetic molecule on a some kind of a molecular leg, will most certainly decrease the tunneling spitting* Δ.

The rigid coupling to the atomic environment is also necessary to provide a small decoherence rate. According to Eq.(28) *the softening of the solid matrix with respect to shear deformations, that results, e.g., in the decrease of c_t by a factor of four, increases Γ by a factor of one thousand.* For $S = 10$ and $c_t \sim 10^5 cm/s$, one obtains from Eq.(28) $\Gamma \sim 10^{-2} s^{-1}$ at $\omega_c \sim 10^{10} s^{-1}$ and $\Gamma \sim 10^3 s^{-1}$ at $\omega_c \sim 10^{11} s^{-1}$. These numbers show that magnetic entities can, in fact, be promising candidates for qubits. It should be emphasized that Eq.(28) establishes the lower bound on the decoherence rate. Other effects unaccounted here, like interaction with nuclear spins, with free electrons, etc., can bring the decoherence rate up.

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